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13. ABSTRACT ***(Maximum 200 Words)***

This DEPSCoR project demonstrated the practical feasibility of ultrafast holographic image interferogram using a single shot exposure with sub-picosecond laser pulses. This goal was achieved by applying novel technique of femtosecond pulsed illumination and by optimizing the recording properties of special organic frequency-selective materials. Recording of single shot femtosecond holograms was accomplished by switching the spectral hole burning material into a photochemically unstable form, which increased the hole burning efficiency by more than one order of magnitude. Double exposure image interferograms were demonstrated with 150-fs duration pulses. Our results show that it is possible to record femtosecond time-resolved holograms and interferograms of ultrafast transient events without loss of spatial/temporal resolution, in a way which overcomes infinitesimal coherence length of the ultrashort pulses.

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ULTRAFAST HOLOGRAPHIC IMAGE RECORDING BY SINGLE SHOT FEMTOSECOND SPECTRAL HOLE BURNING

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Present DEPSCoR grant was closely coordinated with the objectives of a concurrent regular AFOSR grant F4920-98-1-0422, which studied ultrafast holographic image recording by single shot femtosecond spectral hole burning. The work performed under the present grant was focused on the implementation of the holographic and interferometric experiments, whereas the second (regular) grant was focused on optimization of the spectral hole burning material used for recording of the holograms. For this reason, sections of the present summary concerning personnel, lab equipment and publications are partially overlapping with corresponding sections of the final report of AFOSR grant F4920-98-1-0422 submitted one month earlier this year. The main body of the present report describes details of the holographic and interferometric experiments, not covered by any other report.

1. Summary of Results

1.1. Personnel.

The success of this project is largely due to our ability to attract qualified research associates and able students. In 1999 we hired a **Ph.D. scientist** *Dr. Mikhail Drobizhev* , originally from the world-renown Lebedev Physical Institute in Moscow. His specialty is in photophysics and photochemistry of low-temperature organic impurity solids. He is an expert in organic spectral hole-burning materials. Dr. Drobizhev was supported, in part, by this grant. During the period of this project I had **two graduate students**. The first student worked until he fulfilled requirement for a Master's Degree and left for a position in IT-related industry. The second student has passed successfully qualification for Ph.D., and continues to working in my group. Both students have been trained in advanced techniques of femtosecond lasers, ultrafast holography, low-temperature spectroscopy and photochemistry.

Equipment

Project we have essentially built up a new femtosecond laser laboratory starting from scratch. At the end of this grant period, the laboratory has a well-functioning femtosecond laser system with ample wavelength tuning capability. The system consists of a picosecond master femtosecond oscillator (Coherent Mira900) pumped by an Coherent Verdi 5 Watt CW frequency-doubled Nd:YAG laser. The pulses are amplified 1 kHz rate to 0.9 - 1.0 millijoule energy by a Ti:sapphire regenerative amplifier (Clark MXR CPA-1000), which was installed in summer 1998. An important improvement of the laser system was replacing the Ar-ion pump laser (Coherent Innova 400-25) with the solid-state pump laser. Big advantage of the solid state laser is that it has a much better beam pointing stability, and also a better TM00 spatial mode structure for better focusing on the Ti:sapphire crystal. To achieve necessary wavelength tuning in infrared and in visible, we have purchased and installed an optical parametric amplifier (Quantronix Topas). For hologram detection, a cooled high-resolution CCD camera has been purchased and installed.

Part of the matching funds for this DEPSCoR grant provided my the Montana State University were used to buy further equipment such as Molectron pulse energy/power meter, Stanford Research digital high voltage supply, National Instruments GPIB PCI interface card and LabView software, Jobin Yvon CCD array detector and Newport Co. linear actuator and controller.

As a result of this continuing effort, the laboratory has become world class equipped for further experimental work in the broad areas of ultrafast nonlinear optics, femtosecond- as well as linear optical spectroscopy of organic materials.

1.3. Results of the Experiments

1.3.1. SHB materials for femtosecond holographic recording

We started our experiments by addressing the most difficult problem, which consists in finding a frequency-selective material with greatly increased efficacy of spectral hole burning by femtosecond pulses. Our approach was to increase the quantum yield of the process of photo-tautomerization in organic dye-doped polymer material at low temperature. For this we investigated SHB in the unstable photo-tautomer form of some selected porphyrin- and naphthalocyanine molecules. The advantage of using the unstable tautomer form, as opposed to

the conventional stable tautomer form, is that the photochemical back reaction often achieves a much higher quantum yield than the forward reaction. Our final report on the accompanying grant F4920-98-1-0422 gives a detailed account of the spectroscopic investigations we have performed of various SHB recording materials. In particular, we found that best material for the demonstration experiments described below was based on photo-tautomerization back-reaction in anthraceno-tri-phthalocyanine ($AnPc_3$). We have performed detailed spectroscopic studies of this material. Below we briefly summarize the key properties of this most successful SHB material. (a) In polymer matrices, electronic transitions of organic molecules are known to have a large inhomogeneous width, on the order of several THz. The inhomogeneous width of the working $S_1 \leftarrow S_0$ purely electronic transitions of AnPc3 in polyvinylbutyral (PVB) was, $\Gamma_{inh} \sim 7.4$ THz , and in polyethylene (PE) , $\Gamma_{inh} \sim 4.2$ THz . In our experiments we used 150-fs bandwidth-limited pulses with the spectral width (FWHM), $\Delta\nu_p \sim 2.5$ THz. Thus the main requirement that Γ_{inh} exceeds the spectral width of the laser pulses was fulfilled for both polymers. An advantage of PVB over PE is that the first can be hot pressed to give an optically transparent sample, whereas the last exhibits sizable spurious scattering due to microcrystalline structure of the polymer. For reasons described below, we still preferred the PE matrix. (b) In $AnPc_3$ the $S_1 \leftarrow S_0$ purely electronic transitions is electric-dipole allowed, and has a large oscillator strength, $f > 0.01$. It turns out that even for an allowed transition like this, the energy requirement for per single illuminating pulse is quite high: about 100 μJ per cm^2. Furthermore, it in our experiments we established that the sample can dissipate only a finite amount of heat energy per pulse, about 10mW per cm^2. Using higher powers were causing the sample temperature to increase well over 4K, which led to erasure of stored information. The large value of the oscillator strength is also important because creating a coherent excitation in non-allowed transitions such as in rare earth ions on a femtosecond time scale would require a prohibiting high peak power. At liquid helium temperature, $T < 5K$, AnPc3 in polymer matrix has a narrow homogeneous zero-phonon line width, $\Gamma_{hom} < 1$ GHz and favorable large Debye-Waller factor, $\alpha_{DW} \sim 0.7$. (c) The quantum yield of forward photo-tautomerization reaction of AnPc3 is relatively small, $\phi < 0.001$, which is not sufficient for single shot recording. However, we have shown in our experiments, that by illumination at the maximum of the $S_1 \leftarrow S_0$ purely electronic transition at wavelength 780nm, the molecules can be switched into unstable tautomer form, which has the $S_1 \leftarrow S_0$ transition at 764nm. This tautomer form can be switched back to the stable form with a quantum efficiency ϕ

> 0.01, which is already enough for our demonstration experiments. All other key parameters, such as Γ_{hom}, Γ_{inh} and f remain unchanged. The sample used in the experiments consisted of a 0.1-mm thick PE film doped with $AnPc_3$ molecules at a concentration of about 10^{-3} mol/liter. We use $AnPc_3$ in PE matrix because the narrower inhomogeneous width in this polymer allows us to better separate the absorption bands of the two tautomer forms. There are, however, several drawbacks in using polyethylene. First, this polymer doesn't dissolve practically in any solvents, which makes sample preparation extremely difficult. Secondly, polyethylene inherently contains tiny micro crystals, which scatter light and making hologram detection more difficult. To reduce the scattering, the sample needs to be hot pressed to a very thin film (< 0.1 mm). This, in turn, makes sample mechanically fragile and prone to cracking, especially after a few cooling cycles between room temperature and liquid helium. The fact that even after hot pressing procedure, the PE film still comprised an inclusions of small micro-crystals, resulted in a certain level of spurious scattering background. We estimated that because of the micro-crystals, about 10^{-4} of the read beam was scattered in the object beam direction even with no hologram present. Compared to standards used in conventional holography, this is a very high value. Although PVB sample had a far superior optical quality (less scattering), due to twice as large inhomogeneous broadening, there was substantial overlap of the absorption band of the two tautomer forms.

The results of these material studies were reported at several meetings including the 5th International Meeting on Spectral Hole Burning and Related Spectroscopies (HBRS'99), which took place in France in September 1999.

1.3.3. Single-shot hologram recording experiment

Our main result consists in the first time demonstration of the recording of femtosecond image hologram by illumination with a single pair of high intensity femtosecond pulses. This was accomplished by using experimental setup shown in figure 1. The experimental arrangement comprised a regenerative-amplified Ti:sapphire laser (Clark MRX CPA-1000), which produced at the wavelength of λ=764 nm pulses with energy 0.4 mJ and duration of 150 fs at a repetition rate of 500Hz. . The spectral bandwidth of the laser was $\Delta\lambda$ = 5 nm (1 nm wavelength band width corresponds to 0.5 THz in frequency band width). The output laser beam was

expanded to 2 cm diameter and only the central flat intensity profile part was used. A 50% beam splitter divided the beam into the object and reference beams. The two beams were overlapped

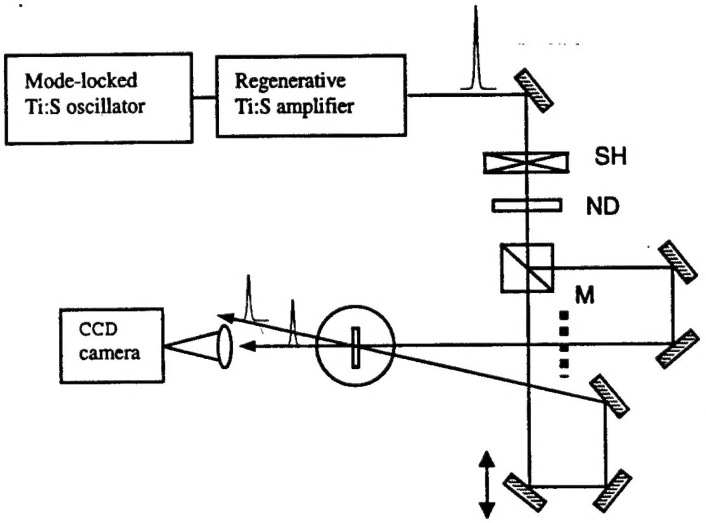

Figure 1. Basic experimental set-up. SH- shutter; ND – neutral density filter; M – transmission mask.

at angle of $\Theta = 5^0$ on the sample, which was positioned inside optical cryostat. The object beam was 10 ps delayed with respect to the reference beam. At the sample, both beams had a diameter of 1 cm and their combined average energy per 1 cm^2 area was 0.15 mJ per laser pulse. The hologram recording exposure was controlled by means of a mechanical shutter which opened and closed the output beam of the amplifier, thus selecting individual pulses form the 500 Hz pulse train. During the experiment the polymer film was immersed in super-fluid helium at temperature T = 2 K. For read-out of the holograms, the object beam was blocked and the reference beam was attenuated by a factor 10^4 with a neutral density filter. Images were recorded with a cooled CCD camera (Xillix Microimager).

Results shown in Figure 2 present the measured dependence of the relative diffraction efficiency of the hologram on the number of single shot exposures. Every single shot exposure contained a pair of object and reference pulses with a median intensity 150μJ cm^{-2}. We estimate that each such exposure burned out about 0.1% of the absorption band at 764 nm. The first thing to notice here is that the diffraction efficiency increases quadratically with the exposure. This can be explained by the fact that the amplitude response of the SHB grating increases linearly with the amplitude of the burnt-in spatial-spectral structure, which, in turn, is linearly proportional to the change of the absorption. Because measured intensity corresponds to the square of the amplitude response, we observe the quadratic increase of the diffraction efficiency with the number of the

applied pulses. The lowest data point in Figure 2 corresponds to an image hologram, which was recorded by two consecutive single shot exposures. The corresponding absolute diffraction efficiency was about $2x10^{-5}$. The background scattering signal due to the imperfection of the PE sample (scattering level 10^{-4} of read beam intensity) has been subtracted from this data.

In addition to the qualitative explanation given above, a simulation based on numerical solution of Maxwell-Bloch equations (solid curve) also predicts nearly quadratic dependence of the relative diffraction efficiency on the number of exposures.

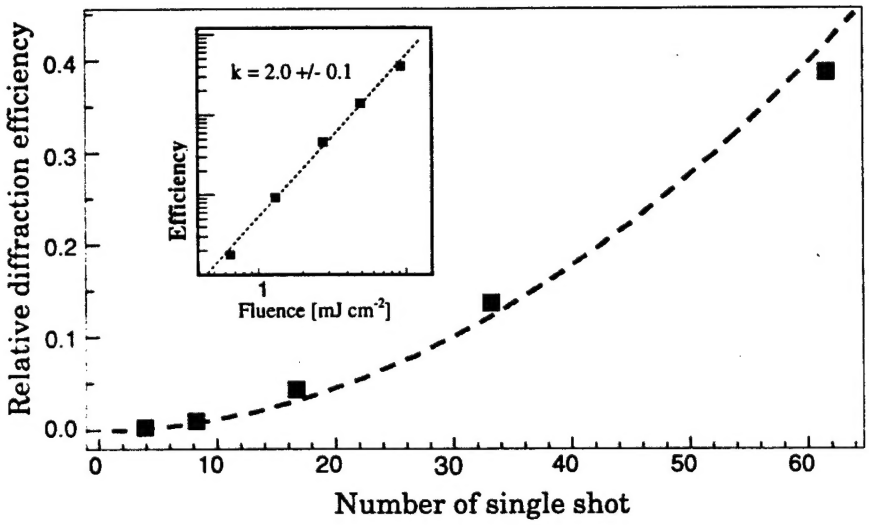

Figure 2. Dependence of relative diffraction efficiency on the number of single shot exposures. Dashed line – fit to numeric simulation yielding a close to quadratic dependence. Insert – the same data on double logarithmic scale.

The smallest absolute diffraction efficiency, at which hologram images were still clearly detected was about $2x10^{-5}$. Taking into account the noise caused by the background scattering, we were able to reliably detect holograms resulting from one or two exposures.

Figure 3 shows actual hologram images recorded in this experiment, and illustrates further the effect of quadratic increase of the diffraction efficiency with the number of exposures. Although holograms recorded with a single shot exposure were detected by eye, their absolute intensity level in CCD images was comparable or less than that of the scattering background noise, and is not presented in this figure.

There are two main conclusions that can be drawn from these experiments. (1) Our results show that by using special frequency-selective light-sensitive materials, it is indeed possible to record femtosecond time-resolved holograms of ultrafast transient events. Furthermore, ultrafast

holograms can be captured without loss of spatial/temporal resolution, which is usually an evitable consequence of infinitesimal coherence length of the ultrashort pulses. (2) Because of steep (quadratic) dependence of the diffraction efficiency on the SHB exposure, one has to use SHB material which combines high optical quality (low background scattering) and high efficency of the photochemical transformation at low temperature. In our case, recording of single shot holograms was accomplished by switching the spectral hole burning material into a photochemically unstable form, which increased the hole burning efficiency by more than one order of magnitude. However, the quality of the hologram images suffered from high scattering background inherent to this particular material. We estimate that an improvement of the detection sensitivity more than two orders of magnitude could be achieved my eliminating this spurious scattering effect.

An advanced report on these results was given at AFOSR Contactors Meeting at SRI International in Menlo Park, CA, May 25-26, 2000.

In this way, the principal objective of this project has been achieved.

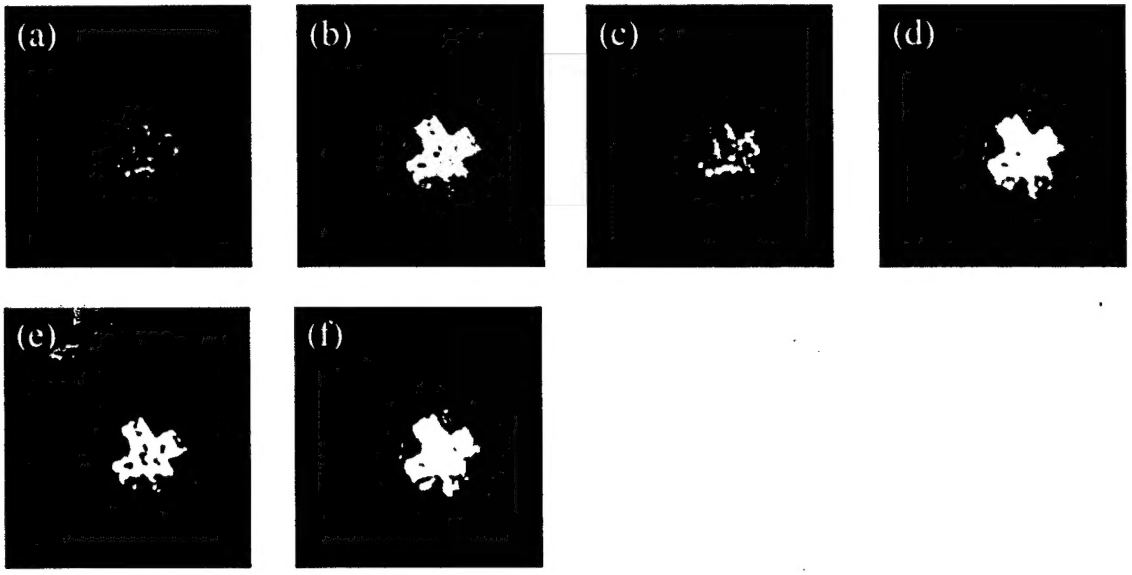

Figure 3. Holograms recorded by accumulating N single shot exposures. (a) N = 2; (b,c) N = 4; (d,f) N = 8; (g) N = 16. Attenuation of the read beam was 1x for (a,b), 2x for (c,d) and 3x for (e,f).

The above results were published, in part, in *Optics Letters* and also reported at several international meetings (CLEO, Physics of Quantum Electronics, etc.).

ometric read-out of femtosecond holograms

nethod of recording femtosecond holograms, we have developed a new
techniques, which can be used to measure the amplitude and phase characteristics
ve forms. In our demonstration experiments, we recorded two spatially-
pping time-space holograms: the first hologram contained the object wave pulse subject under investigation; the second holograms contained a reference wave front with a known plain amplitude and phase structure. By illumination of the hologram with a plain wave pulse, both stored wave fronts are reproduced simultaneously. However, because the time delay of the two reproduced pulses with respect to the read pulse are not necessarily the same, we don't observe any interference between these two recalled amplitudes. However, if we read out the hologram with a narrow-band light, at a wavelength corresponding to one spectral component of the pulse, then the two hologram signals do overlap, and we observe interference between the object and the reference waves. Such composite hologram is an analog to double-exposure hologram well known in conventional holographic interferometry. The novel property of this method is that it can be used to capture simultaneously the time-domain as well as space-domain (image) interferogram of the object pulse. Figure 3 shows an example of a double-exposure femtosecond interferogram. First, we recorded a relatively strong plane wave reference hologram by accumulating 80 single shot exposures using 150-fs pulses of the mode-locked Ti:sapphire laser.

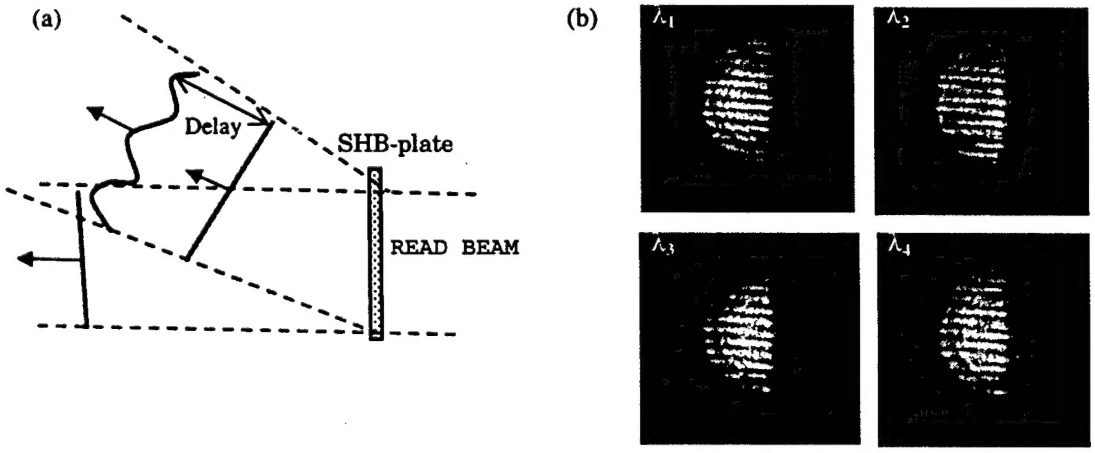

Figure 4. Double expose femtosecond holographic interferograms. (a) Scheme of readout with plane wave read beam. The object wave diffracts from the SHB-plate in the same direction as time-delayed, plane wave front reference wave recorded in a different exposure. The time delay between the object and the reference is achieved by changing the delay of the writing beam (b) Images observed in the object bema direction at four different wavelengths of the readout laser. Note the difference of the position of the interference fringes.

After that we exposed the sample by using one laser shot, but with a slightly tilted reference beam direction and with a slightly different delay. Left side of Figure 4 shows the two wave fronts, which were reproduced from the hologram if it was illuminated with femtosecond read pulse. Note the time delay between the pulses, which prevents interference between the two amplitudes. We then switched the Ti:sapphire oscillator to CW operation. In the CW regime, the spectral line width of the laser was less than 0.1 nm, and the wavelength was tuned by Lyot filter element in the cavity. Right hand side of Figure 4 shows interference patterns observed as the readout wavelength was varied. This figure shows that spatial phase of the object pulse is different at different frequencies, or, in other word, the interferogram is indeed revealing full spatial-spectral phase structure of the object pulse.

Although the demonstration achieved in our present experiments was rather primitive, we believe that this approach can be perfected to give eventually a useful practical tool for investigating ultrafast transient phenomena.

1.3.5. Patents and Commercialization Activities

We have started a new collaboration with Dr. Charlie Spangler from Montana State University Chemistry Department on application of organic chromospheres with large two-photon absorption cross-section. The principal idea is to use these materials for detection of mode-locked infra-red laser pulses, and potentially also as a memory material for optical storage. We have filed a patent "Mode-Locked Laser Infrared Detection Card and Method" (US Patent application 09/834,727). This technology is a licensed to a laser materials company in Bozeman, Scientific Materials Co.

2.3.6. Conclusion

We have accomplished the principal goal of this project: we have performed an experiment which shows that capturing a holographic image of a unique ultrafast event on subpicosecond time scale in a single recording exposure is indeed practically possible. This was achieved, on one hand, by investigating spectral hole burning properties of a range of organic materials, and on the other hand, by carefully optimizing the experimental conditions. Successful demonstration was achieved by modifying a previously known spectral hole burning materials in way, which increased the recording efficacy by more than one order of magnitude. This allowed

us to record image holograms with 150-fs duration pulses without need to accumulate the SHB effect from many exposures. Results of this research show that it is possible to perform optical recording of data in frequency-domain on ultrafast time scale. These results can be used also as a new diagnostic tool for femtosecond dynamics in various ultrafast optical interactions.

3. Publications

3.1. Articles in refereed journals

1. "Terahertz, bit-rate parallel multiplication by photon echo in low-temperature dye-doped polymer film," O.Ollikainen, C.Nilsson, A.Rebane, Optics Commun. 147 (4/6), 429 (1998).
2. "Ultrafast time-and-space-domain holography in dye-doped polymers," A.Rebane, Chimia 52, 112-117 (1998).
3. "Nondestructive read-out of two-color photon-gated spectral hole burning holograms," D.Reiss, A.Rebane, U.P.Wild, Mol.Cryst. Liq. Cryst, v.314, 161-166 (1998).
4. "Photo-tautomer of Br-porphyrin: a new frequency-selective material for ultrafast time-space holographic storage," M.Drobizhev, C.Sigel and A.Rebane, J. of Luminescence 86, 391-397 (2000).
5. **"Single femtosecond exposure recording of image hologram by spectral hole burning in unstable tautomer of phthalocyanine derivative," A.Rebane, M.Drobizhev, and C.Sigel, Opt. Lett. 25, 1633-1635 (2000).**

3.2. Conference proceedings and presentations

1. A.Rebane, "Organic frequency-selective materials for ultrafast optical storage and processing," SPIE Proceedings, v.3468, 270-278 (1998).
2. A.Rebane, "Organic materials for ultra-fast holographic storage and processing," Optical Science and Laser Technology Conference, Bozeman, August 17-18, 1998.
3. A.Rebane and D.Reiss, "Nondestructive and transient read-out of photon-gated hole-burning holograms," CLEO, San Francisco, May 1- 4 1998.
4. O.Ollikainen, J.Gallus, U.Wild, A.Rebane, "New coherent optical technique for single-shot homogeneous spectrum measurement," CLEO, San Francisco, May 1- 4 1998.
5. O.Ollikainen, J.Gallus, C.Nilsson, D.Erni, A.Rebane, "Terahertz bit-rate optical processing by femtosecond two-pulse photon echo," CLEO, San Francisco, May 1- 4 1998.
6. Workshop on Spectral Hole Burning, Bozeman, March 8-11, 1998.
7. (INVITED) A.Rebane, SPIE 43rd Annual Meeting, Conference on Advanced Optical Memories and Interfaces to Computer Storage, 19-24 July 1998, San Diego.

8. (INVITED) A.Rebane, Topical Meeting of the International Commission for Optics "Optics for Information Infrastructure", Tianjin, China, 3-6 August 1998;

9. Workshop on Spectral Hole Burning, Bozeman, March 1999.

10. (INVITED) A.Rebane, 12th International Conference on Dynamical Processes in Excited States of Solids, Humacao, Puerto Rico, May 23 - 27, 1999.

11. A.Rebane, "Efficient frequency- selective materials for ultrafast time- and frequency domain data storage and processing", 6th International Meeting on Hole Burning and Related Spectroscopies: Science and Applications, Hourtin, France, September 18-23, 1999.

12. A.Rebane, "Femtosecond holography and pulse interactions in inhomogeneously broadened media," 30th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, Utah, 9-12 January 2000.

13. A.Rebane, M.Drobizhev and Ch. Sigel, "Single-shot recording of ultrafast time-space holograms," CLEO 2000, San Francisco.

14. A.Rebane, M.Drobizhev and Ch. Sigel, "Photon-gated holographic hole burning and read-out in Si-naphthalocyanine-doped polymer film," M.Drobizhev, C.Sigel, and A.Rebane, CLEO 2000 Proceedings, p.488 (2000).

15. A.Rebane, "Interference between femtosecond pulses observed via time-resolved spontaneous fluorescence," Workshop on Applications of Spectral Hole Burning, Big Sky, July 9 -12, 2000.

16. A.Rebane, "Femtosecond holography and pulse interactions in inhomogeneously broadened media," 30th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, Utah, 9-12 January 2000;